Comment on "Adiabatic stabilization: Observation of the surviving population"

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Abstract

Questions are raised about certain experimental and theoretical claims that atoms may be stabilized into their bound states, and prevented from achieving full ionization, by the application of adiabatic, ultraintense, high-frequency laser pulses. It is pointed out that those authors have used the weak-field concepts of cross section and ionization rate in an ultra intense field regime where they have no physical significance.

The purpose of this Comment is to raise several questions concerning the theoretical interpretation of results obtained from recent experiments by van Druten et al. [1]. In those measurements Ne atoms in a cell were prepared in the $(2p)^5 5g$ $m = 4$ Rydberg state by a pumping laser and then ionized by a 90 fs pulse of 620 nm laser radiation having the range of peak intensities $.05 \times 10^{14}$ to 2.3×10^{14} W/cm^2. The relative yields of ions thus produced and the surviving 5g atoms were measured. Figure 8 in Ref. [1] contains the results of these relative yields, normalized so they represent the absolute probabilities P(ion) and P(5g). Any appreciable leakage into other channels was assumed negligible, so that the authors converted their relative measurements to absolute ones with the normalization $P(ion) + P(5g) \cong 1$ at all intensities. Their $P(ion) \cong .25$ and is virtually flat between $I = .5 \times 10^{14} W/cm^2$ and $2.3 \times 10^{14} W/cm^2$, and the authors point out that this behavior is well below the ionization probability expected on the basis of the Fermi Golden Rule. They also claimed that the relative closeness of their extracted ionization cross sections to those of the theory of Potvliege and Smith [2] constituted a verification of the existence of "adiabatic stabilization."

We should like to point out to the reader that a dispute has been going on over the past decade on whether or not "adiabatic stabilization" does indeed exist. Unfortunately, such controversy was not acknowledged in Ref. [1]. Briefly, the proponents of this phenomenon claim that when using sufficiently high-intensity, high-frequency fields, the ionization rate may decrease as the field intensity increases. Since this is a clearly unexpected and counter-intuitive behavior, it is not surprising that it is a matter of considerable theoretical controversy. For a detailed presentation of the arguments,

we refer the reader to the general review articles by Eberly and Kulander [3] in favor of stabilization, and by Geltman [4] in favor of full ionization. These review articles contain extensive reference lists.

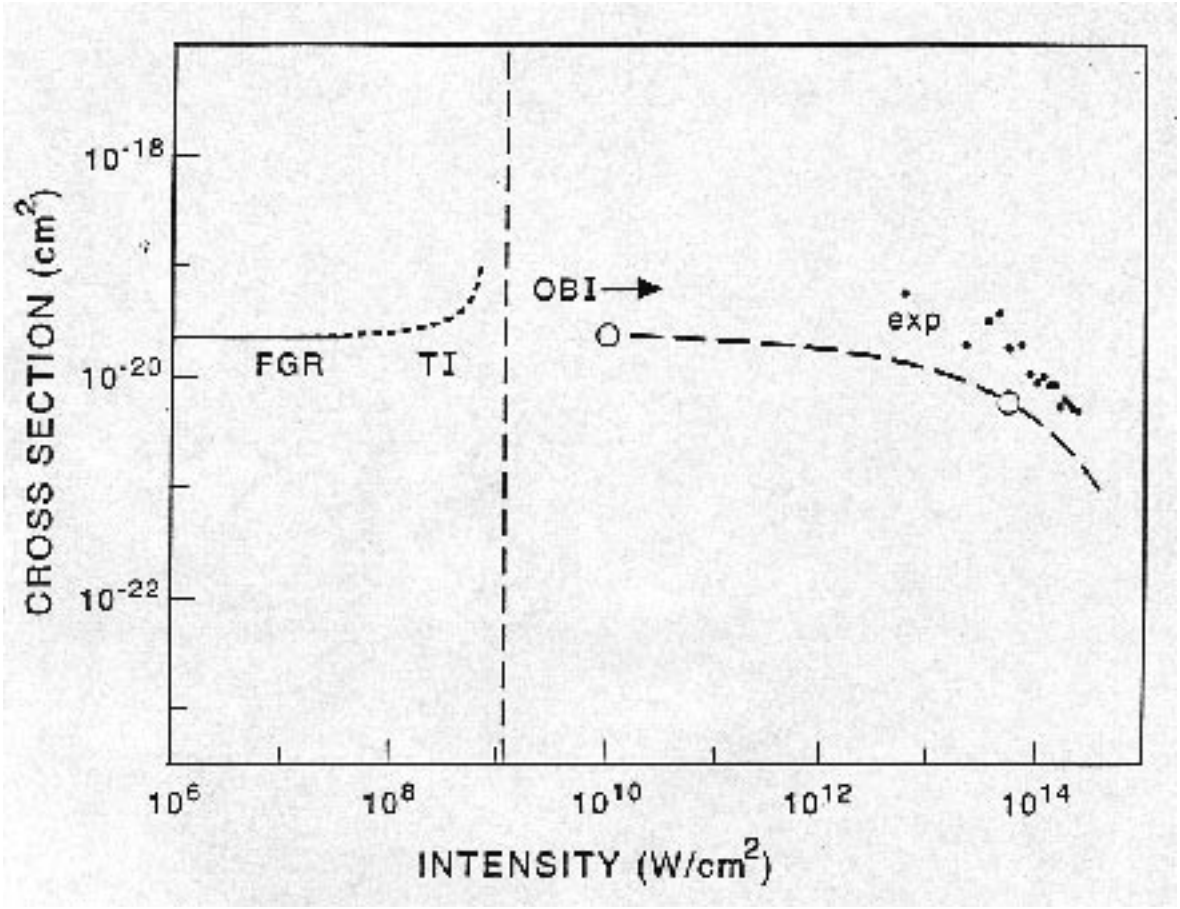
The overall situation as applied to the experiment [1] and theory [2] under discussion is shown in Fig. 1, where the one-photon cross section for the ionization of $Ne(5gm = 4)$ is plotted as a function of laser intensity. For the purposes of the present discussion the atom $Ne(5gm = 4)$ may be regarded as purely hydrogenic. The 16 points for P(ion) given in Fig. 8 of [1] are represented here by the points labeled "exp." The theoretical predictions at high intensity in Fig. 1 are those of Potvliege and Smith [2] who use a Sturmian-Floquet method to evaluate lifetimes of various hydrogenic states subject to very intense fields at a number of wavelengths. In general they find this lifetime to decrease when the field intensity increases, until a minimum in lifetime occurs, followed by an increase in lifetime when further increasing the intensity. We believe that this unexpected behavior is most likely the result of the use of a theoretical method beyond its range of validity. The Sturmian-Floquet method cannot account for the time dependence contained in the full time-dependent Schrödinger equation under such extreme conditions, where the atomic binding potential is effectively destroyed by the applied field.

It is central to this discussion at this point to indicate the dilemma that one faces when trying to understand how these results connect to the picture of photoionization at much lower field intensities, where there is general agreement. At the lowest intensities the Fermi Golden Rule (FGR) allows the evaluation of exact one-photon photoionization cross sections, and one finds that $\sigma\ (5gm = 4) = 2.15 \times 10^{-20} cm^2$ for $\lambda = 620nm$, using a method of Burgess [5]. This value is indicated by FGR at the far left of Fig. 1. At higher intensities one expects corrections over lowest-order perturbation theory to enter the picture, and for tunneling (TI) to become the dominant mechanism for ionization. At even higher intensities the "tunnel" disappears as the effective binding potential falls below the level of the bound state. This occurs for a hydrogenic atom (m=0) when

$$E_o = Z^3/16n^4, \tag{1}$$

where E_o is the peak electric field in the laser pulse (in a.u.), Z is the effective nuclear charge, and n is the principal quantum number. This field strength is the threshold for a very rapid and nonlinear rise in the probability (to essentially unity) of the bound electron escaping into the continuum,

Figure 1: Effective cross section of the photoionization of the hydrogenic $5g$ $m = 4$ state by 620 nm laser radiation as a function of intensity. The FGR calculated value is indicated at the lowest intensities (solid line), as are the approximate regions for the FGR, TI, and OBI modes of ionization. The dashed line in the TI region is a schematic representation of the cross section below the OBI threshold (vertical dashed line). Experimental points of van Druten et al. [1] for Ne $5g$ $m = 4$ (filled points) and the calculated values of Potvliege and Smith [2] (open circles and approximate connecting curve).



over the lowered barrier along the field direction, called over-the-barrier ionization (OBI). For the present 5g state and Z=1, one finds that this condition is reached at a laser intensity of $3.51 \times 10^8 W/cm^2$. Cooke and Gallagher [6] have pointed out that when $m \neq 0$ there is a correction to the threshold field value given by (1) which is due to the preservation of the kinetic energy associated with the angular momentum along the quantization axis. The inaccessibility of that energy to the translational kinetic energy of the ejected electron amounts to a raising of the effective threshold field of OBI. Making that correction we find that the OBI intensity for the $5g$ $m = 4$ state is raised to $1.18 \times 10^9 W/cm^2$. The above three regions are indicated by FGR, TI, and OBI in Fig. 1. The effective cross section in the TI region has not been evaluated, but only schematically indicated as the dashed line rising from the FGR limit to reflect the physically expected increasing cross section with decreasing tunneling barrier size.

For all intensities above the OBI threshold an effective cross section may not be meaningfully defined since the ionization probability is no longer a linear function of the time. We must therefore regard as anomalous the experimental [1] and theoretical [2] points above the OBI threshold in Fig. 1. In the TI and OBI regions the more precise quantity to describe the ionization dynamics is the ionization probability, which results from a particular laser pulse. The basic mechanisms for electron ejection in these regions are tunneling and field emission, which are qualitatively different from that of photon absorption, the mechanism that applies at the lowest intensities. The proper description in the TI and OBI regions requires the solution of the full time-dependent Schrödinger equation followed by its projection onto field-free continuum states, a task that has so far been intractable for real atoms. Calculations on model atoms [7] have shown the rapid rise to full ionization, as expected from the above qualitative arguments, and rigorous deductions of the absence of stabilization have also been given [8]. The use of an ionization rate or bound state lifetime, as done in [2], no matter how sophisticated, can at best describe the true physics only through the TI region. Above the OBI threshold the ionization probability is no longer a linear function of the pulse duration, and so no ionization rate is any longer physically meaningful.

It is very difficult for a reader who is not actively engaged in similar experiments to pinpoint exactly where erroneous results may have arisen in this experiment [1]. An absolute measurement of ionization probability as presented in Ref. [1] would require the perfect alignment of three lasers to ensure that they all are acting on the identical interaction volume in the cell.

For example, it is not clear that the axial dimensions of all the focal regions are identical, which would cast doubt on whether all the $Ne(5g\ m=4)$ atoms produced by the preparation pulse were exposed to the peak intensity of the ionizing pulse. Furthermore, the measurements of relative yields of surviving atoms and ionized atoms may not be reliably converted to absolute probabilities by the simple normalization used in Fig. 8 of Ref. [1]. One must have individual absolute probability measurements for each of these yields to ensure that no other channels are interfering. Ideally, an accurate absolute measurement of ionization probability would require the use of crossed atom and laser beams rather than a gas cell, as it is such crossed beam geometries that have provided the most accurate measurements in the past.

In conclusion, the purpose of this Comment is to show the gulf between a phenomenological understanding of photoionization in ultra-intense fields and the measurements reported in Ref. [1] and the theory in Ref. [2]. These reported results on "adiabatic stabilization" use the concept of ionization rates by photon absorption in an intensity region 3 to 5 orders of magnitude larger than that at which one would expect the total break-up of the atom by simple electrostatic arguments. To become acceptable, such claims for stabilization must provide a physical answer to the counter argument that the atom is being completely dissociated at much lower intensities. How can there be any appreciable surviving population when the top of the binding potential barrier lies far below the bound-state energy? There is no reason that we can see to expect any appreciable survival population for a bound atomic state that has been subject to many ($\sim$50) cycles of a field of such extremely high intensity.

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